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ARPA-NBS Program of Research on High Temperature Materials and Laser Materials

Reporting Period
January 1 to June 30, 1971

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ARPA-NBS Program of Research on High Temperature Materials and Laser Materials

Reporting Period

January 1 to June 30, 1971

Edited by A. D. Franklin and H. S. Bennett

Inorganic Materials Division
Institute for Materials Research
National Bureau of Standards
Washington, D.C. 20234

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ARPA-NBS PROGRAM OF RESEARCH ON HIGH TEMPERATURE
MATERIALS AND LASER MATERIALS

Work Performed at the National Bureau of Standards
Supported by the Advanced Research Projects Agency,
Department of Defense
(ARPA Order 373-62)

Reporting Period January 1 to June 30, 1971

Edited by
A. D. Franklin and H. S. Bennett

Progress reports are given for projects on the growth of ultra-pure Al_2O_3 crystals, the development of a mass spectrometer-based sectioning technique for measuring oxygen diffusion in oxides and the development of a precision facility for measuring the threshold energy in a laser beam producing damage in a transparent substance.

Key words: Al_2O_3 ; crystal growth; damage threshold; glass; high temperature materials; laser; oxides; oxygen diffusion; pure materials; sapphire.

1. INTRODUCTION

The National Bureau of Standards, with the support of the Advanced Research Projects Agency (Office of Materials Sciences) of the Department of Defense, is carrying out a program of research on High Temperature and Laser Materials. The overall objective of this program is to provide some of the key elements needed by DoD in its pursuit of new technology through research and development. These elements include:

- i. The preparation and characterization of materials needed for critical studies where exacting criteria of chemical purity, crystalline perfection, etc. must be met if progress is to be made,
- ii. The development of new measurement techniques where these are required, and the acquisition of critically-needed data on certain material properties, and:
- iii. The elucidation of the basic mechanisms contributing to or limiting the use of materials under extreme conditions (e.g. high temperatures, high intensity laser light, etc.).

A summary of the results achieved in the period January 1 to June 30, 1971, is given here.

Among the many areas of Defense Technology where advances of the kind described above are needed, this Program has focussed upon two. Ceramics are used in many ways, and many problems in their use relate ultimately to mass transport effects. Highly important improvements could be made by developing a firmer understanding of and control over mass transport processes in oxides. Since NBS has had considerable experience in this field, it is a natural one upon which to concentrate a portion of this Program.

The other area of concentration was introduced into this Program several years ago in response to an urgent need seen by the Sponsor. As laser power has moved upward, laser beams, especially pulsed beams of very short pulselength, have been found to damage the materials (ampli-

fier laser rods, windows, lenses, prisms, and other optical elements) through which the beams must pass. Not only must a better understanding be developed of the processes leading to the several types of damage so far observed, but also there is a need for more reproducible techniques for studying these damage processes and for characterizing the response of given materials with respect to them.

Corresponding to these two areas of concentration, this Report is divided into two parts, High Temperature Materials and Laser Materials. Each part will open with a brief summary of objectives and accomplishments, followed by detailed Project progress reports.

2. HIGH TEMPERATURE MATERIALS PROGRAM

2.1. Objectives and Summary of Program in High Temperature Materials

2.1.1. Objectives

The operation of rocket and jet engines involves exposing metals at high temperatures to oxidizing conditions, and raises the problem of oxidation protection of high temperature refractory metal structural components. This same problem, currently for superalloy turbine blades, is posed again by jet engines. The oxidation protection of metals requires some form of barrier layer to prevent diffusion. This layer, either applied or developed by the early stages of oxidation itself, will most likely be an oxide, and will function to keep the metal and oxygen apart. It can work only if the diffusion rates of oxygen and metal atoms through the layer are small enough. Thus, the strategy for the development of oxidation protection of otherwise susceptible metals at high temperatures will depend in part on being able to control the rates of diffusion, and therefore, in having data on the diffusion rates themselves.

Diffusion in oxides is important to other classes of DoD problems. Thus improvements in the reliability and efficiency of military communication, computer, and radar circuitry could be made if ceramic magnetic and ferroelectric components could be made more reproducible.

and reliable. Because such important properties as initial susceptibility and coercive force of ferrites, and dielectric and piezoelectric properties of ceramic ferroelectrics, are very sensitive to the microstructural features (porosity, grain size) controlled by sintering, the strategy for improvement here requires control over sintering in these ceramics. The same is true of ballistic response of ceramic armor. The development of transparent armor requires new techniques for the suppression of grain growth during otherwise complete sintering of inherently transparent ceramics like Al_2O_3 .

Since sintering in ceramics is mainly brought about by diffusion processes, the development of successful, long-range strategies to meet these DoD needs rests in part on knowledge of the rates of diffusion of metal and oxygen atoms in oxides.

Reliable techniques for measuring oxygen diffusion rates are not yet available, and therefore reliable data for oxygen diffusion does not exist for any save a few oxides. Thus one objective of this program is to develop a reliable technique for measuring the rate of diffusion of oxygen in oxides.

Further, diffusion rates are notoriously sensitive to impurities, and strategies for control require knowledge of the basic rate in the pure material and of the way various classes of impurities influence it. This in turn requires very pure crystals for study. To date, almost no oxide crystals have been prepared pure enough to reveal the intrinsic diffusion rates. The second objective in this area is to prepare intrinsically pure crystals of those materials for which mass transport data are sorely needed. The first candidate has been Al_2O_3 , an important constituent in many ceramics, in particular for ceramic armor.

2.1.2. Summary of Progress to Date

In previous work a vapor deposition technique had been perfected for the growth of Al_2O_3 single crystals, and the resulting crystals

shown to be very pure. Sources of residual impurity were shown to be mainly the metal parts of the growth apparatus exposed to hot gases and the starting Al metal.

A revised equipment has been constructed and will be used, together with the purest obtainable Al metal to attempt the growth of ultra-pure Al_2O_3 crystals for intrinsic mass transport measurements.

A key problem in developing the equipment for measurement of the oxygen diffusion coefficient in oxides has been the conversion of the oxide sections to a form suitable for mass spectrometer analysis. During the past period an apparatus for the chemical decomposition of the sections by BrF_5 has been built and is being tested. The reaction has been mainly successful, although the yields of oxygen for the mass spectrometer analysis have been lower than anticipated and desired. The explanation may lie in contamination of the starting BrF_5 . Additional work will be done during this next period to maximize this yield, and it is also planned to begin diffusion studies on MgO crystals.

2.2. Project Summaries in High Temperature Materials

2.2.1. Diffusion in Oxides

2.2.1.1. CRYSTAL GROWTH FROM VAPOR

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The objectives of the program are, first, the growth of aluminum oxide mono- and bicrystals of sufficient physical perfection and chemical purity for use as research materials, second, the reduction of both cation and anion impurities to sufficiently low levels to permit meaningful property measurements at the intrinsic level and third, the extension of the technique to other materials of interest.

During the present reporting period, the growth of high purity mono- and bicrystals has continued, in order to supply additional

specimens to other ARPA investigators concerned with mass transport in oxides. Relocation of the growth apparatus to a different laboratory caused some interruption to the experimental work. An additional modification to the growth apparatus has been the inclusion of an oil-wetted mechanical filter between the liquid nitrogen cold trap and the vacuum pump to prevent amorphous alumina powder remaining in the gas stream from reaching the pump. This should materially extend the life of the pump.

The components for power and temperature control of the new system described in previous reports have been received and wired and the new system will be in operation within two weeks. It is expected that the combination of the new growth system with a minimum of exposed metal parts to hot gases and the use of 99.9999 per cent pure aluminum metal source material will produce specimens of the highest purity attainable by this technique.

A recalculation of the magnetic susceptibility of our specimens has shown an error in the previously reported value of $0.318 \pm 0.002 \times 10^{-12}$ m³/g.¹

¹ NBS-ARPA Report for the period January - June 1970, A. D. Franklin and H. S. Bennett, Eds., NBS Report No. 10 353, p.9.

The corrected value is $0.360 \pm 0.002 \times 10^{-12} \text{ m}^3/\text{g}$, in much better agreement with the theoretical value of $0.38 \times 10^{-12} \text{ m}^3/\text{g}$, as would be expected for material of this purity.

During the next reporting period it is planned to complete the growth and characterization of high purity single crystals in the new apparatus and complete the preparation of a publication describing the technique and results.

2.2.1.2. DIFFUSION IN REFRACTORY MATERIALS

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The objectives of this program are to measure and to interpret diffusion of oxygen in single crystal oxides. Experimentally these two objectives consist of (1) extending the sensitivity of present techniques so that the ^{18}O distribution in a single crystal sample can be measured, (2) comparing the results of these concentration profile measurements with measurements of the rate of exchange of ^{18}O between the gas phase and the oxide crystal, (3) determining the dependence of the diffusion rate on temperature, oxygen partial pressure, impurities in the solid, etc. and (4) obtaining reliable diffusion and exchange coefficients. Oxygen self-diffusion in oxides provides a tool for uncovering the type of point defects involved in ionic migration in oxides and the energies required for the formation and motion of these defects. Since oxygen diffusion can be the controlling step in the solid state reaction and sintering of oxides, the diffusion coefficients are needed for interpretation and estimation of these rates.

One of our experimental problems is to determine the ^{18}O distribution in oxide crystals after a diffusion anneal. To do this we are endeavoring to couple sectioning, i.e., removal of layers of about

1 μm in thickness from the crystals, to analysis of the " $^{18}\text{O}/^{16}\text{O}$ " ratio with a mass spectrometer. The chemical reaction which provides this coupling is the most difficult step in the procedure. Preliminary results from the reaction of BrF_5 with MgO , Al_2O_3 and TiO_2 have encouraged us to believe that at last we may have the chemical tool which will accomplish this coupling.

The free energies for the reactions of BrF_5 with Al_2O_3 , MgO and TiO_2 are favorable even at room temperature (1)

<u>Oxide</u>	<u>$\Delta G^\circ_{298.15}, \text{ kJ}$</u>
Al_2O_3	-603
MgO	-151
TiO_2	-426

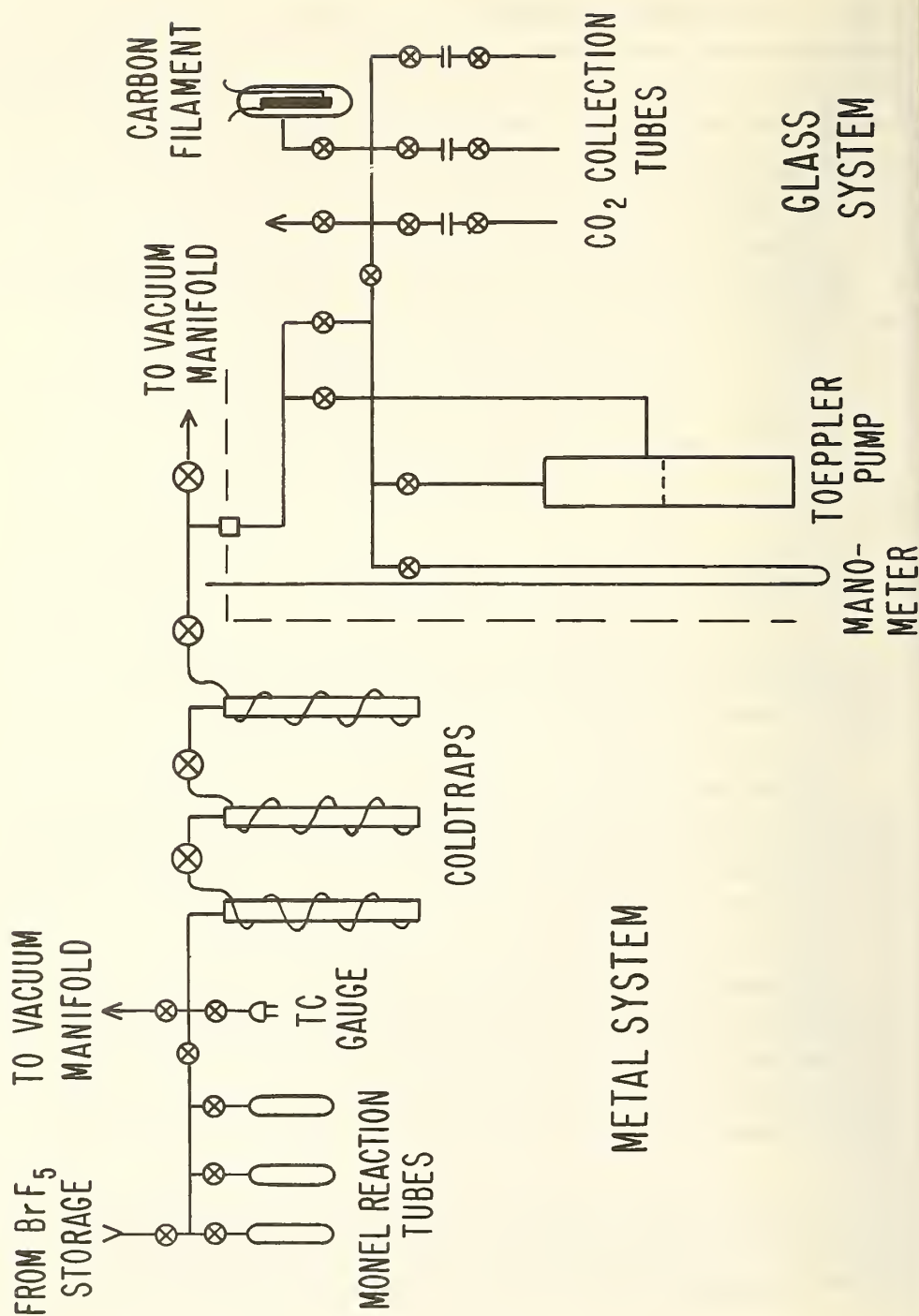
The products are assumed to be BrF_3 , O_2 and the fluorides of the respective metals. BrF_3 also will react with the oxides.

Ruff and Menzel(2) were the first researchers to report on the reaction of BrF_5 with a number of oxides. Since that time geochemists (3) have developed procedures for its use in oxygen isotopic analysis of water and of silicate rocks.

Our experimental arrangement is detailed schematically in the accompanying drawing. The "Metal System" is used to purify and to store the BrF_5 , to react the BrF_5 with oxide samples in the monel reaction tubes and to separate the toxic gaseous fluoride products from the oxygen in a chain of three liquid nitrogen-cooled coldtraps.

The "Glass System" is used to collect the oxygen, to measure the quantity of oxygen obtained, to convert the O_2 to CO_2 using a hot carbon filament and to collect the CO_2 for analysis with the

BrF₅ - OXIDE REACTION APPARATUS



mass spectrometer. Carbon dioxide is more desirable for isotope ratio measurements than is oxygen.

We estimate the yields from our initial experiments to be

20 - 30% for MgO

60% for Al_2O_3

65% for TiO_2 .

In current experiments we are measuring the amounts of reactants and products with greater care, purifying the BrF_5 and investigating the influence of KF on the reaction(4).

References

1. JANAF Tables, Dow Chemical Company, Thermal Research Laboratory.
2. O. Ruff and W. Menzel, Z. anorg. allg. Chem., 202, 49 (1931).
3. J. R. O'Neil and S. Epstein, J. Geophys. Res., 71, 4955 (1966).
4. I. Sheft, A. F. Martin, and J. J. Katz, J. Amer. Chem. Soc., 78, 1557 (1956).

3. LASER MATERIALS PROGRAM

3.1. Introduction to Laser Materials

The National Bureau of Standards, with support from the Advanced Research Projects Agency (Office of Materials Sciences) of the Department of Defense, is performing research on laser materials. This research includes studies to characterize laser materials and to define and evaluate damage measurements in laser materials. A summary of the results achieved during the reporting period is given here.

3.2. Objectives and Summary of NBS Research on Laser Materials

3.2.1. Objectives

Past experience in the high-power laser field indicates a need to improve the reproducibility and precision of measurements concerning damage and degradation in laser materials. As in other fields, this requires an understanding of the important material and instrumental

characteristics and their influence on the results of the measurement. Such a requirement for laser damage measurements means having a well-characterized laser beam in the sense that we know and can vary the intensity as a function of position, time, and frequency for each pulse.

To this end, we have constructed and are putting into operation a neodymium-doped glass laser system as the fundamental measurement tool. We are taking great care in characterizing and controlling the beam. The objectives of this program then include:

1. To study the measurement of the threshold for surface and for self-focussing damage, the statistical nature of the measurement, and the influence on it of the beam characteristics; and
2. To study on a microscopic-physical and chemical basis the nature of surface and intrinsic damage in various materials. These studies will probably include:
 - a. The influence of electrostrictive and Kerr effects on self-focussing and intrinsic damage; and
 - b. The effect of surface preparation and history on surface damage.

Even though the failure of laser materials due to optically-induced damage still hinders somewhat the advance in high power laser technology, a significant change in the approach to the problem occurred this past year. Whereas up to a year ago researchers considered that particulate damage was the major problem, they now consider that intrinsic mechanisms (e.g., self-focussing, surface damage, and plasma formation) should receive the greatest attention.

The recent symposium at Boulder rather dramatically illustrated this change. Speakers at this Third Symposium on Damage in Laser Materials (May 1971) mentioned only briefly the particulate (extrinsic) damage problem in Nd-doped glass, a problem of great concern just one year ago. This reflects more a success in technology than an advance in science. It is now possible to make glass with

so few particles that they are apparently no longer the primary source of damage. Surface damage, the dynamics of the processes leading to surface damage, the statistical aspects of damage and degradation processes, and the damage to laser materials for high power applications at 10.6 μm have become now some of the foremost problems to occupy the efforts of researchers. This latter group of problems is associated more with the intrinsic damage processes than with the extrinsic damage processes due to impurities whose concentrations may be reduced to acceptable levels.

Past experience suggests that the intrinsic damage problems will not yield as easily to the technological approach as did the particulate problem. We may solve the latter by removing the particles, but in order to ameliorate the intrinsic damage mechanisms, we very probably must first develop the science of intrinsic damage; that is, we must understand the damage processes and their relations to material characteristics. This development of the science underlying the intrinsic damage problem will require us to make proper, well-controlled measurements of the damage threshold for each mechanism as we seek correlations with changes among material compositions and treatments. These are the reasons why the NBS program has adopted the objective outlined above.

3.2.2. Summary of Progress to Date

A theoretical model for studying the thermal stresses produced by inclusions in laser materials has been developed. A paper containing the numerical predictions of this model was published during the period covered by this report.

i. H. S. Bennett, Absorbing Centers in Laser Materials, J. of Applied Physics, 42, 619 (1971).

The results of past projects on the characterization of laser materials also were published during this period. These projects have been terminated.

ii. R. M. Waxler, Laser Glass Composition and the Possibility of Eliminating Electrostrictive Effects, IEEE J. of Quantum Electronics,

Q. E. 7, No. 4, 166 (1971).

iii. R. M. Waxler, et al., Optical and Mechanical Properties of Some Nd-doped Laser Glasses, NBS J. of Res., 75A, No. 3, 163 (1971).

A commercial Nd-glass laser is being modified for precise single-mode operation. Its oscillator section now operates satisfactorily, and its performance characteristics are being determined with precision. In addition, a Twyman-Green interferometer and a polariscope have been constructed. They are used to test the optical properties and quality of all optical components in the laser.

The modified oscillator has been used for preliminary studies on the intrinsic damage associated with self-focussing in borosilicate crown glass.

The data from such studies agree reasonably well with the data of others and include values of wavelengths and pulse widths which other researchers have not reported. However, the present theoretical models to interpret these data need many improvements. Their numerical predictions do not agree well with the measurements.

During the next period it is planned to improve the two amplifier sections of the laser, to continue in more detail the measurements on damage threshold, and to examine on a microscopic-physical and chemical basis the several damage mechanisms.

3.3. Project Summaries in Laser Materials

3.3.1. Damage to Laser Materials

3.3.1.1. Laser Induced Damage Studies

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I. Objectives

The overall objectives for the laser materials program are discussed in Section 3.2.1. Briefly, they are to measure thresholds for surface and intrinsic bulk damage and to examine on a physical and chemical basis the nature of such damage. The objectives for the present reporting period are more limited. They are first to operate the oscillator section of our laser in a well-controlled, single-mode (TEM_{00}) and second to study the mechanism of self-focussing and the intrinsic damage which may be produced by self-focussing.

II. Approach

Self-focussing is a precursor to damage from high power laser beams. Preliminary measurements of the critical power for self-focussing have been made in one material and compared with existing data and theories. A method was found for comparing data obtained with lasers using different wavelengths and pulse widths. When our data are scaled, they agree favorably with previous measurements. Further measurements are planned at increased power levels by use of two amplifiers. Higher power levels are necessary for testing current theories of self-focussing.

III. Laser System

The major part of this report period has been concerned with the construction of a Nd:glass laser test facility. This involved ordering replacement parts for the laser system, setting up a Twyman-Green interferometer for the testing of all optical components, and performance tests on the laser oscillator.

The original laser system has been described previously(1). The oscillator has been modified to produce a beam in the TEM_{00} mode by using apertures up to 3.15 mm in diameter. The oscillator rod is 12.7 mm in diameter by 177.8 mm long and has a $6^\circ - 6^\circ$ window configuration.

The unpumped wavefront distortion is less than $\lambda/10$. (All measurements were made with a helium-neon laser operating on the 632.8 nm line.) The output reflector is a 45% dielectric mirror. Q-switching is presently done with a Pockels cell. The two amplifier rods are 19.1 x 304.8 mm and 19.9 x 215.9 mm respectively and have $6^\circ - 6^\circ$ window configurations. The wavelength distortion in each rod is less than $\lambda/4$. All rods are pumped with helical flash lamps.

IV. Testing of Optical Quality

A. Twyman-Green Interferometer

A Twyman-Green interferometer has been set up in order to test the optical quality of all optical components in the system. This gives us the ability to measure flatness and wavefront distortion to within $\lambda/20$. Figure 1 shows a schematic diagram of the interferometer. The light source is a two milliwatt helium-neon laser in the TEM_{00} mode, and is polarized vertically. The power is adequate for measuring wavefront distortion in our longest rods (304.8 mm).

We find that we achieve the best fringe visibility when the optical path lengths in the two arms are equal or when the difference between the optical paths in each arm is an integer multiple of the length of the helium-neon laser (about 43 cm). However, we find that fringe patterns are discernible even when this optical path difference varies by as much as 10 cm. from a given integer multiple. Hence, wide variance in the difference between optical path lengths is tolerable.

B. Effect of Pumping on Oscillator Rod

We performed tests on the oscillator rod while mounted in the head to determine the uniformity of pumping. After carefully aligning the flashlamp to be concentric with the laser rod, we inserted the head in one arm of the Twyman-Green interferometer. The fringe patterns were photographed at less than one second and at several successive times greater than one second after the flash lamp was fired. These are shown in figure 2. The fringe pattern immediately

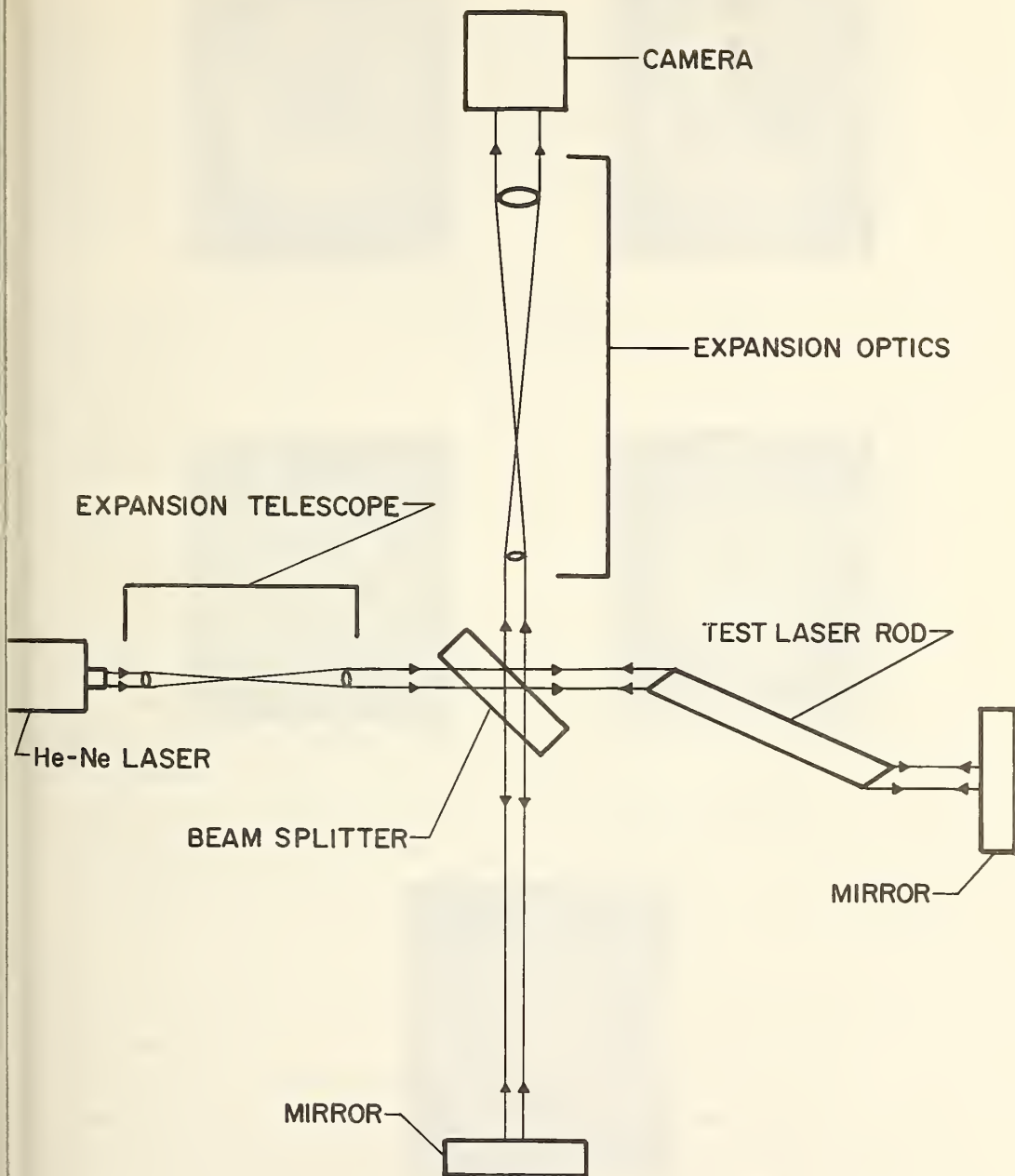
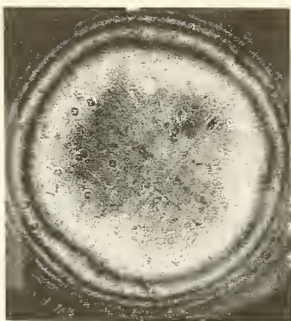
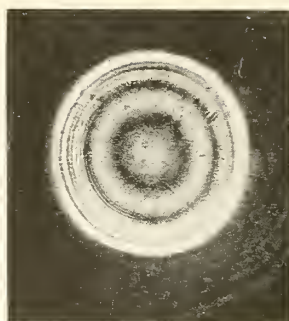


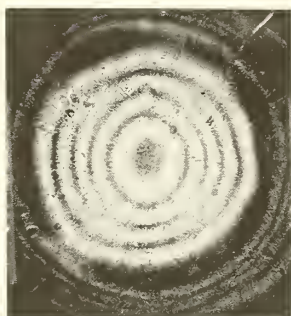
Figure 1. Twyman-Green Interferometer



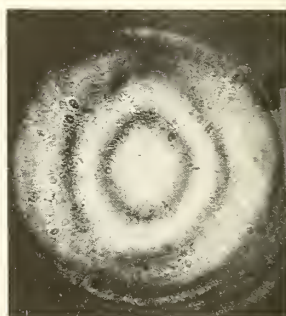
(a)



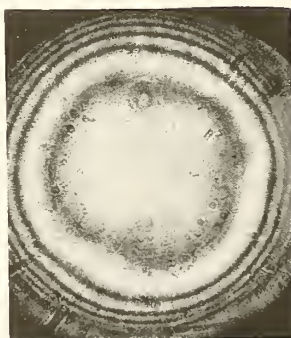
(b)



(c)



(d)



(e)

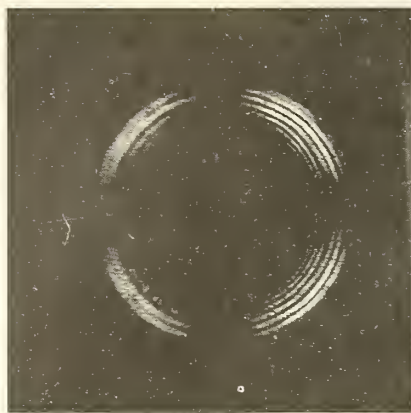
Figure 2. Twyman-Green interferograms of oscillator rod after pumping. (a) Before pumping (b) Less than one second after pumping (c) After 10 seconds (d) 20 seconds (e) 40 seconds.

after the flash is cylindrically symmetric, indicating cylindrically symmetric pumping. The decreased size of the image shows that the rod is acting like a positive lens. From figures 2 and 3 and from other observations we judge that maximum distortion in the rod takes place about 5 seconds after the flash. Essentially all distortion in the rod disappears after 40 seconds.

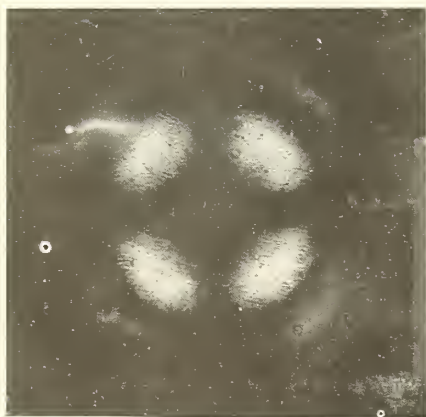
In figure 2c, we observe that the fringe pattern has a two fold symmetry with the fringes disappearing at about 45° . This effect is caused by cylindrical stress in the rod, and the polarization of light. In the 12 and 6 o'clock positions the light is polarized in the radial stress direction, while in the 3 and 9 o'clock positions the polarization is in the tangential stress direction(2). Thus if any stress-induced birefringence is present we would expect to see the two fold fringe pattern.

The presence of pump-induced stress in the rod was observed by blanking off the reference arm of the interferometer and inserting an analyzer in crossed position before the camera. The result is a double pass polariscope. Figure 3 shows the birefringence produced by pumping. The pattern again shows cylindrical symmetry. The cross in the pattern occurs where the principal stress axes are parallel to the light polarization.

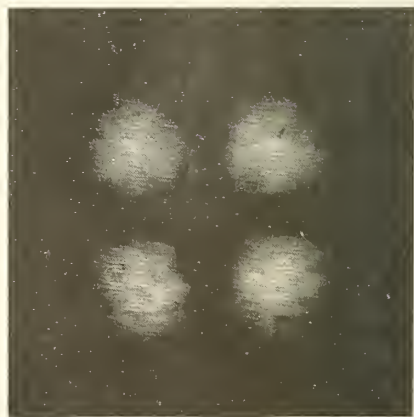
The temporal development of the stresses in the rod agrees with that calculated for stresses in a uniformly heated cylinder suddenly plunged into a constant temperature bath(3). These calculations were made on the basis of the measured parameters of the neodymium-doped glass(4). The fact that it takes a relatively long time (5 seconds) for the maximum stresses to develop indicates that the photograph immediately after the flash is fairly close to that which would appear during the flash. Welling and Bickart(5) have taken high speed photographs which show that stresses are set up during the flash. These stresses are due almost entirely to thermal gradients caused by heating as a function of radius in the rod.



(a)



(b)



(c)

Figure 3. Photographs of oscillator in double pass polariscope after pumping. (a) Less than one second after pumping (b) After 5 seconds (c) After 10 seconds.

C. Pockels Cell Aperture

The oscillator Pockels cell was placed in the polariscope to examine its aperture as a function of applied voltage. Figure 4 shows a photograph of the aperture. Note that the aperture is not in the center of the cell. The maximum size of the aperture is 4.7 mm in diameter. The Pockels cell was placed in the oscillator so that the aperture was centered along the desired beam axis.

V. Oscillator Performance

Tests were made on the oscillator to determine total energy output, cross sectional energy density distribution, time evolution of pulse, and pulse reproducibility. As presently set up, all tests could not be made simultaneously. Figure 5 shows a schematic representation of the test apparatus. All tests were performed under constant oscillator pumping conditions.

A. Pulse Energy Measurements

The total energy per pulse was measured using a thermopile energy meter directly in the path of the laser beam. The manufacturer claims an accuracy of $\pm 5\%$ for the energy meter. Using an aperture of 2.44 mm and a pump energy of 3.53 kJ, we obtained an output energy of 99 mJ with no deviation larger than $\pm 3\%$.

B. Cross-Sectional Energy Distribution

The cross-sectional energy distribution in the laser beam, or beam profile, is the energy H per unit area falling on a surface perpendicular to the beam direction as a function of distance from the beam axis. The beam profile of our laser beam is obtained from burn patterns on developed unexposed black (Polaroid)² film. The output of the laser is maintained at a constant pulse energy but is attenuated from pulse to pulse by interposing calibrated attenuation filters between the oscillator and the film. Figure 6 shows the set of burn patterns used for determining the beam profile. The radius of

²Commercial materials are identified in this paper to specify the particular substance on which the data was obtained. In no instance does such identification imply recommendation or endorsement by the NBS or that the material identified is necessarily the best for the purpose.

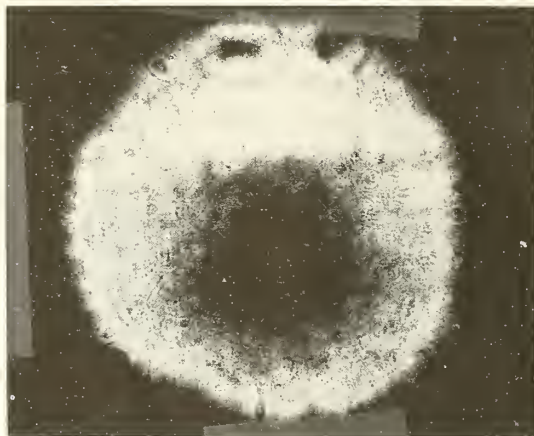


Figure 4. Pockels cell as seen in double pass polariscope with 8 KV applied using the helium-neon 632.8 nm line. The aperture is 4.7 mm.

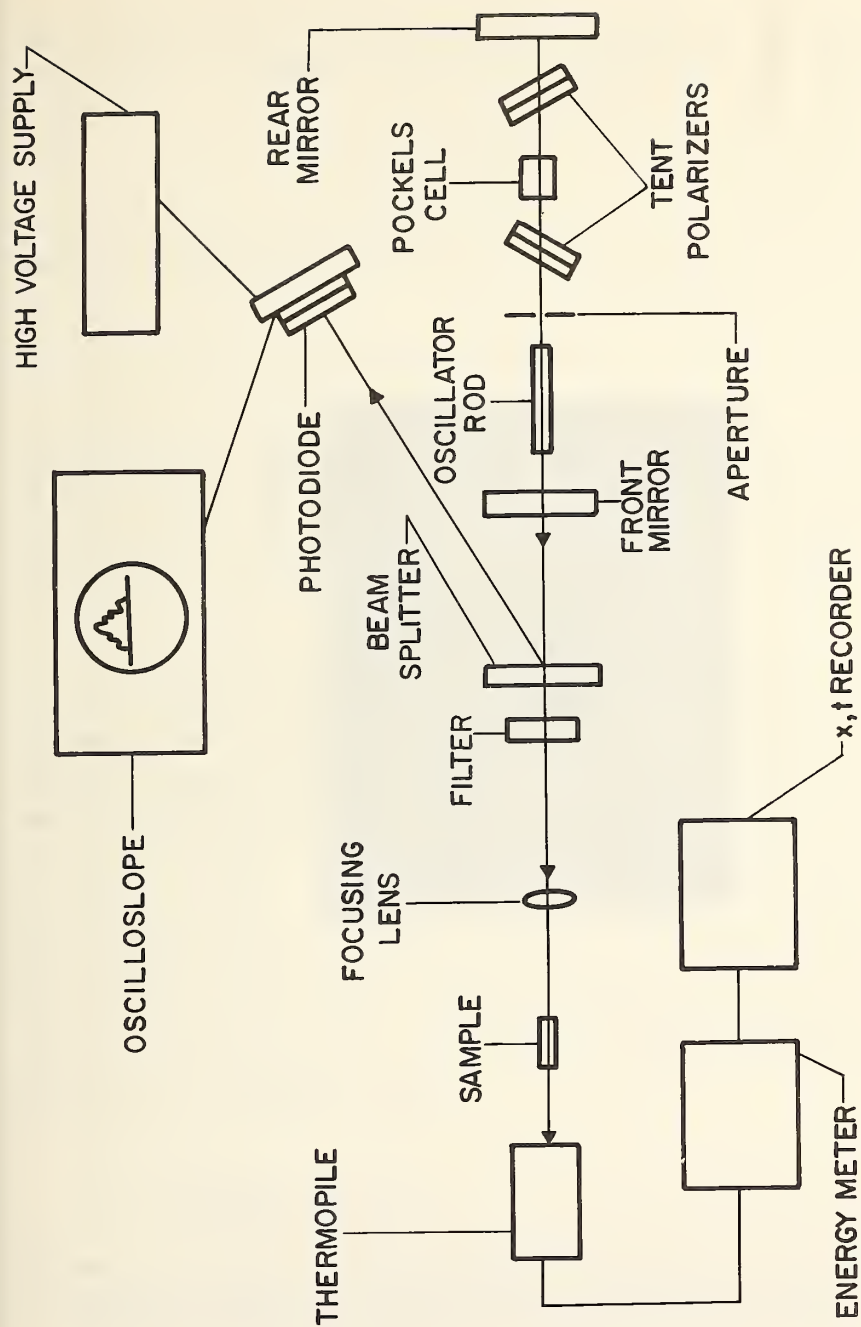


Figure 5. Apparatus for measuring oscillator performance and for damage testing.

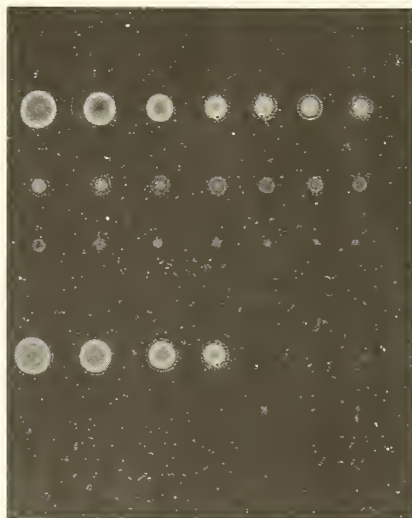


Figure 6. Burn patterns on developed unexposed (Polaroid) film used for determining beam profile at position of focusing lens.

the outermost burn ring for each pattern is plotted as a function of beam energy. The result is fitted to a Gaussian

$$H = H_0 \exp \left(-\frac{r^2}{a^2} \right) .$$

The constant H_0 is obtained in units of J/cm^2 from

$$E = 2\pi H_0 \int_0^\infty \exp \left(-\frac{r^2}{a^2} \right) r dr ,$$

where E is the total pulse energy (99mJ). Figure 7 shows a plot of the beam profile together with the Gaussian which is fitted to the data by eye. The profile, which is measured at a distance of 482 cm from the oscillator aperture, has a $1/e$ point radius, a , of 1.81 mm.

It is important to know the energy distribution at the aperture in the oscillator. For this purpose it is useful to assume that the profile is indeed Gaussian. A Gaussian beam has the property of maintaining a Gaussian shape throughout all of space. At one position it has a minimum width which is called the beam waist. Using the results of Boyd and Gordon(6) we obtain a beam waist diameter ($1/e$ point in the beam profile) of 0.93 mm which is assumed to occur at the oscillator aperture (we neglect the amplification of the oscillator rod in the calculation). The ratio of the aperture diameter (2.44 mm) to the beam waist diameter is 2.62. At the aperture radius, therefore, the intensity of a Gaussian falls to 0.1% of its peak value and contains 99.9% of the total beam energy.

C. Pulse Time Evolution

In order to measure the laser pulse time evolution, three percent of the beam is reflected from a fused silica beam splitter into a high speed biplanar photodiode with a rise time of less than 0.3 nsec.

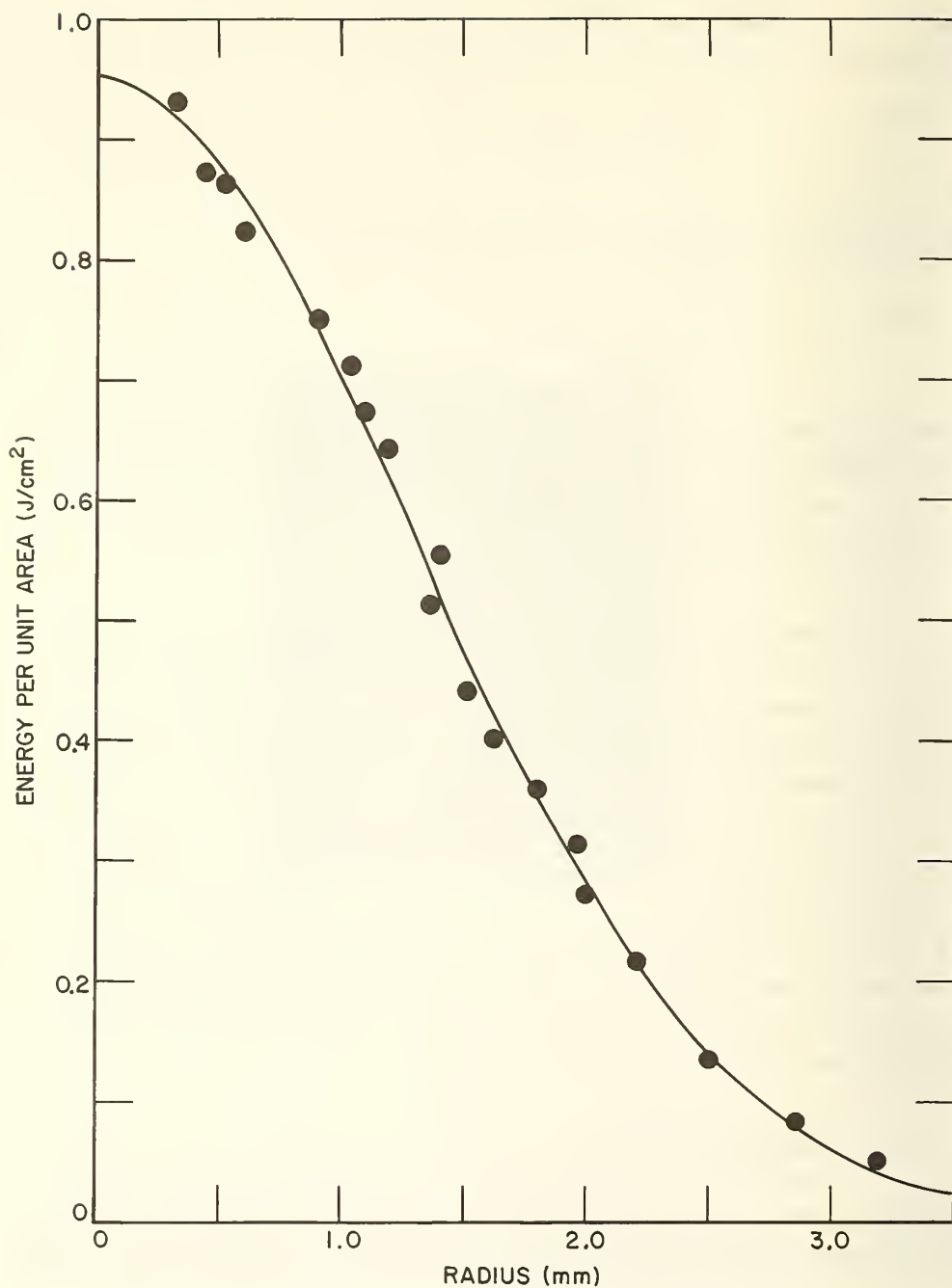


Figure 7. Energy per unit area as a function of distance from laser beam axis as measured at the focusing lens. The solid line is a gaussian fit to the data.

The signal is displayed on a high speed oscilloscope.

Figure 8 is a photograph of the oscilloscope trace. The ripples on the pulse are due to the beating of adjacent longitudinal modes of the oscillator, whose optical path length is about 86 cm. The pulse width is 20 nsec at half the maximum power. By integrating the pulse area using a planimeter, and by knowing the time base and total energy, we obtain a value for the peak power of 4.1×10^6 watts.

VI. Preliminary Self-Focussing and Damage Test

Preliminary measurements of self-focussing and damage in borosilicate crown glass (BSC 517) are made by focussing the laser beam into the sample. The test apparatus is shown in Fig. 5. Table 1 gives all the relevant beam parameters as they are obtained in section V.

A. Test Procedure

The focal point of the focussing lens whose length is nominally 152 mm is determined to within 1.25 mm by taking burn patterns on a special high optical density glass(7) (Schott NG 1 glass). The sample of BSC glass, a rectangular prism 38 x 38 x 76 mm, is placed in the beam so that the square front face was 25.4 mm in front of the focus. Thus, the focus in the glass, whose refractive index is 1.507 at $1.06 \mu\text{m}$, is 38.2 mm from the entrance face.

The laser is fired into the glass and a damage track is observed. The laser beam is subsequently attenuated from shot to shot, until no damage is observed.

B. Results

Figure 9 is the picture of a damage track. The downstream end is predominantly a narrow filament while the upstream end shows catastrophic damage. An end view of the track shows that the fractures are planes intersecting on the track axis. These results are similar to the damage observed by others.(8)

TABLE I
BEAM PARAMETERS
GAUSSIAN BEAM PROFILE

Wavelength	1.06 μm	
Aperture Diameter in Oscillator	2.44 mm	
Beam Energy099 J	
Pulse Width (Full Width Half Maximum)	20 nsec	
Peak Power	4.1×10^6 W	
Aperture to Lens Distance	482 cm	
Lens Focal Length	152 mm	
	<u>At Focussing</u>	<u>At Lens</u>
	<u>Lens</u>	<u>Focus</u>
Peak Energy Per Unit Area (J/cm ²)	0.95	14,600
Peak Intensity (W/cm ²)	4.0×10^7	6.1×10^{11}
Beam Radius 1/e Energy Per Unit Area	1.81 mm	14.6 μm





20 nsec

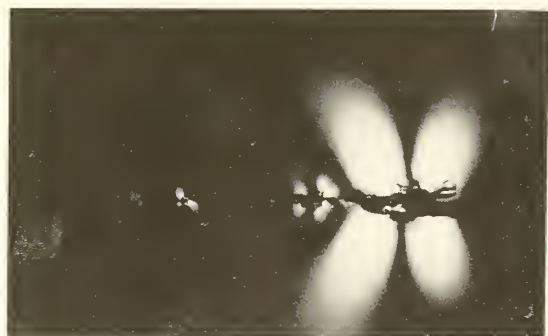
Figure 8. Oscilloscope trace of the time evolution of the laser pulse.



(a)



(b)



(c)

Figure 9. Damage track in BSC 517 glass.

(a) Laser beam direction is from right to left.

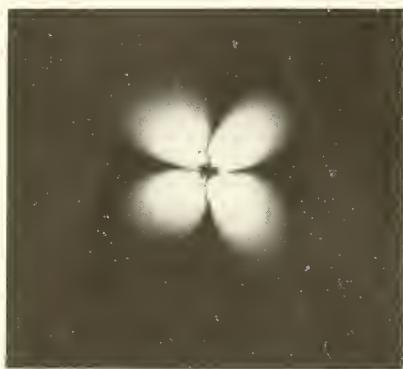
(b) End view of damage track.

(c) Track as seen through crossed polaroids. The pattern observed is indicative of strain associated with the damage sites.

In addition we have a view of the track observed in crossed polaroids. Strain-induced birefringence patterns are observed, with the greatest strains associated with the largest damage sites. The presence of the strains suggests that the glass is heated rapidly to a high temperature and then is cooled rapidly.

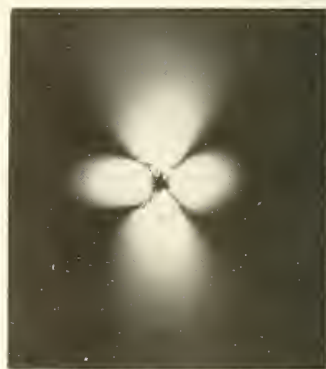
Figure 10 shows the end view of a damage track between crossed polaroids. A cross is observed, as mentioned in section IV B, in directions where the principal stress axes are parallel to the polarizer and analyzer. The second picture (10b) represents a rotation of the polarizers by 45° . If the stress field were radially symmetric, then the stress patterns in both pictures would possess four fold symmetry and would be identical. A slight two fold asymmetry is observed. This could be either an experimental artifact or else could be associated with the polarization of the laser whose beam is polarized predominantly in the horizontal plane. Kerr(9) has shown that one might expect to find effects due to linear polarization in electrostrictive self-focussing because of anisotropy in the electrostrictive forces.

Figure 11 shows the positions of the damage tracks with reference to the front face of the sample and with reference to the expected position of the lens focus. Also given are the lengths of the tracks and the peak power of the laser pulse used to produce the damage. It is known that all tracks start downstream(10). Notice that the lower energy tracks are the farthest downstream. From the theory of self-focussing and from results obtained previously(10), one expects that all damage tracks would start closer to the lens focus for the beam configuration used than was observed. The effect observed is not understood, and will require additional investigation.



┌───┐
4 mm

(a)



(b)

Figure 10. End view of damage tracks in BSC 517 glass between crossed polaroids.
(a) Polarizer parallel to polarization direction of damaging radiation.
(b) Polarizers rotated 45° .

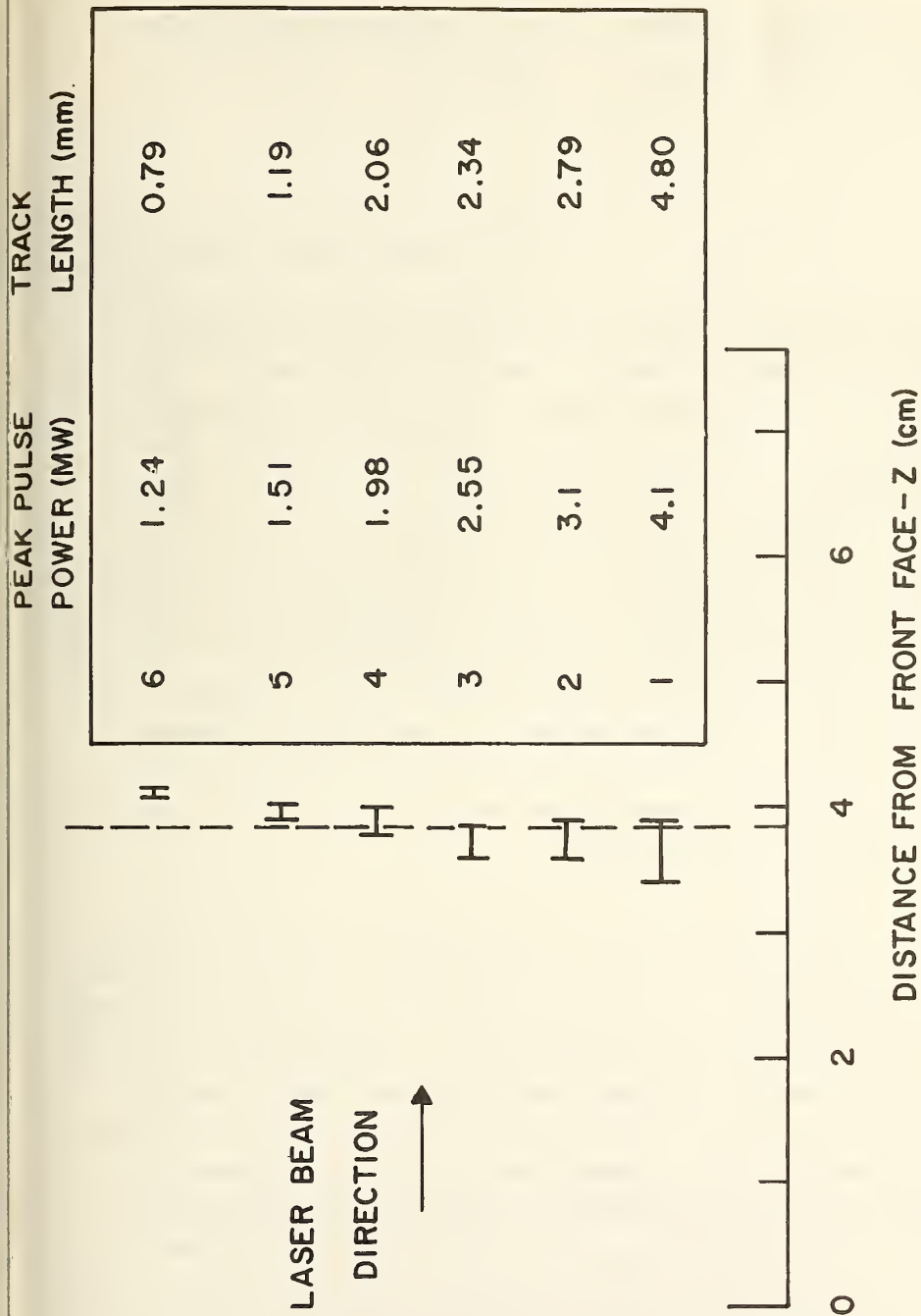


Figure 11. Position of damage tracks in BSC 517 glass with reference to entrance face of sample. Also given are the powers used to produce each track and the track lengths. The dashed line represents the nominal focus of the focusing lens.

An attempt is made to fit the limited data to current theories of self-focussing and to earlier data. A self-focussing length, z , is calculated from the data of figure 11 using the single lens equation(11)

$$z^{-1} = z_f^{-1} - z_c^{-1},$$

where z_f is the distance of the upstream end of the damage track from the sample entrance face and z_c is the threshold self-focussing length for a focussed laser beam which is taken to be the distance from the downstream end of the lowest power track to the entrance face. The inverse self-focussing lengths were plotted as a function of input power. A value for the critical power for self-focussing, $P_c = 1.18$ MW, was obtained by extrapolating empirically to $z^{-1} = 0$. The inverse normalized self-focussing lengths $(z^*)^{-1}$ are plotted in figure 12 together with the theoretically calculated curve of Dawes and Marburger(12) (our P_c is equivalent to their P_2) where $z^* = z/2ka^2$ and $k = 2\pi n_o/\lambda$; n_o is the refractive index in the medium; λ is the wavelength of the laser beam in air; a is the radius of the $1/e$ intensity point at the sample face. The data qualitatively follow the theory, although they do not quite fit the theoretical curve. The theory assumes that the refractive index is

$$n = n_o + n_2 E^2$$

whereas if electrostrictive or thermal self-focussing is important the situation would be more complicated.

Figure 13 shows the threshold power for self-focussing in BSC glass as a function of the laser beam radius at the focus of a lens, where the beam radius a_o is calculated neglecting self-focussing effects. The figure is from the work of Kerr(9) and of Steinberg(13). Also, the data from this report are shown in the box on the figure. The solid circles represent powers at which damage occurred and the open circles represent powers at which no damage occurred. The X represents our threshold value. The solid curve is a theoretical fit from Kerr's theory of electrostriction(9). It is necessary to scale our data because our wavelength ($\lambda = 1.06 \mu\text{m}$) and pulse width ($\tau = 20$ nsec) differed from those of Steinberg ($\lambda = 694.3$ nm, $\tau = 55$ nsec). The theory of electrostrictive self-focussing(9) gives an expression for the self-focussing power threshold

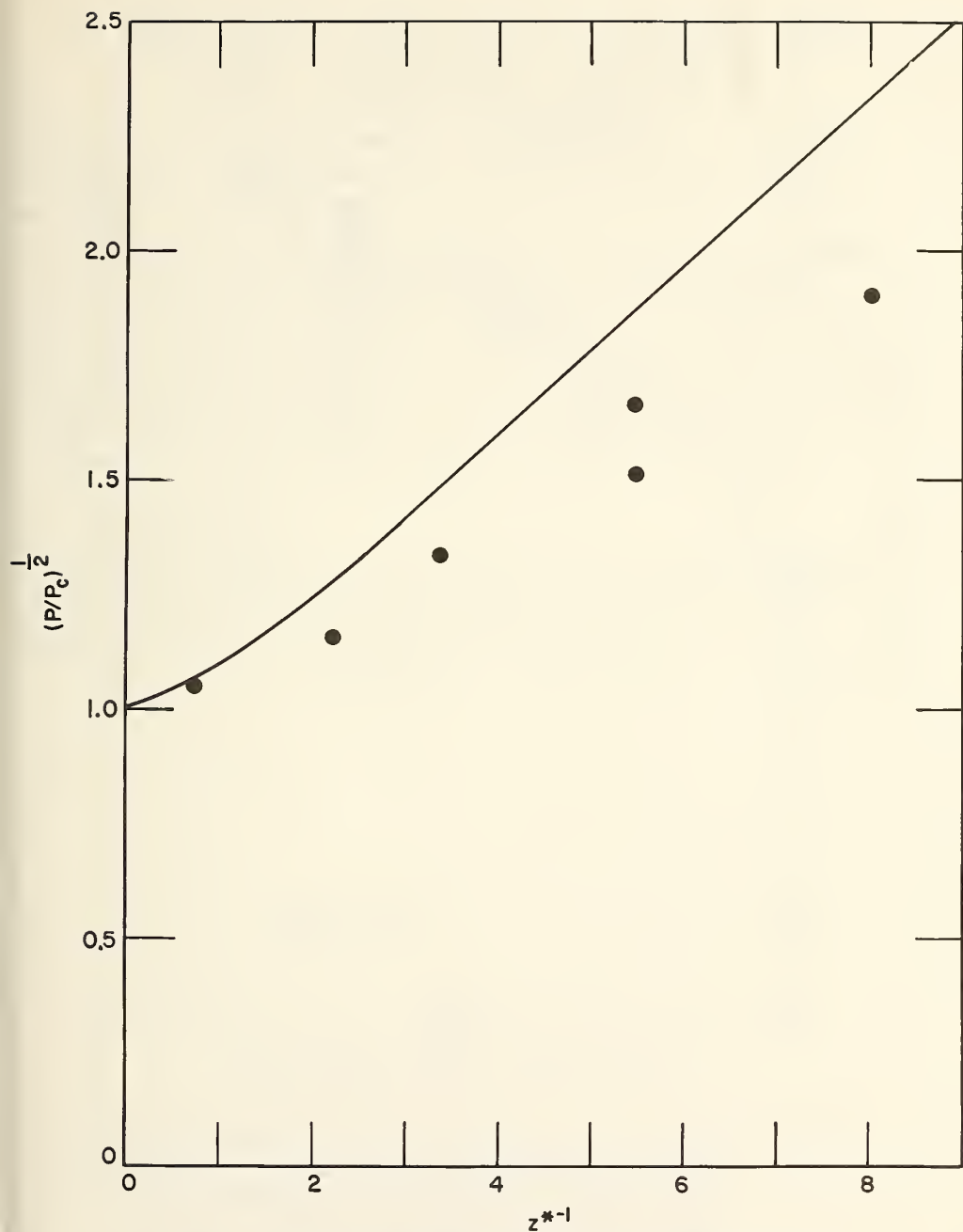


Figure 12. Self-focusing length in BSC glass as a function of beam power. The solid curve is a theoretical curve derived from Dawes and Marburger (Ref. 12). Assumptions made to obtain this plot are given in the text. The critical power $P_c = 1.18$ MW.

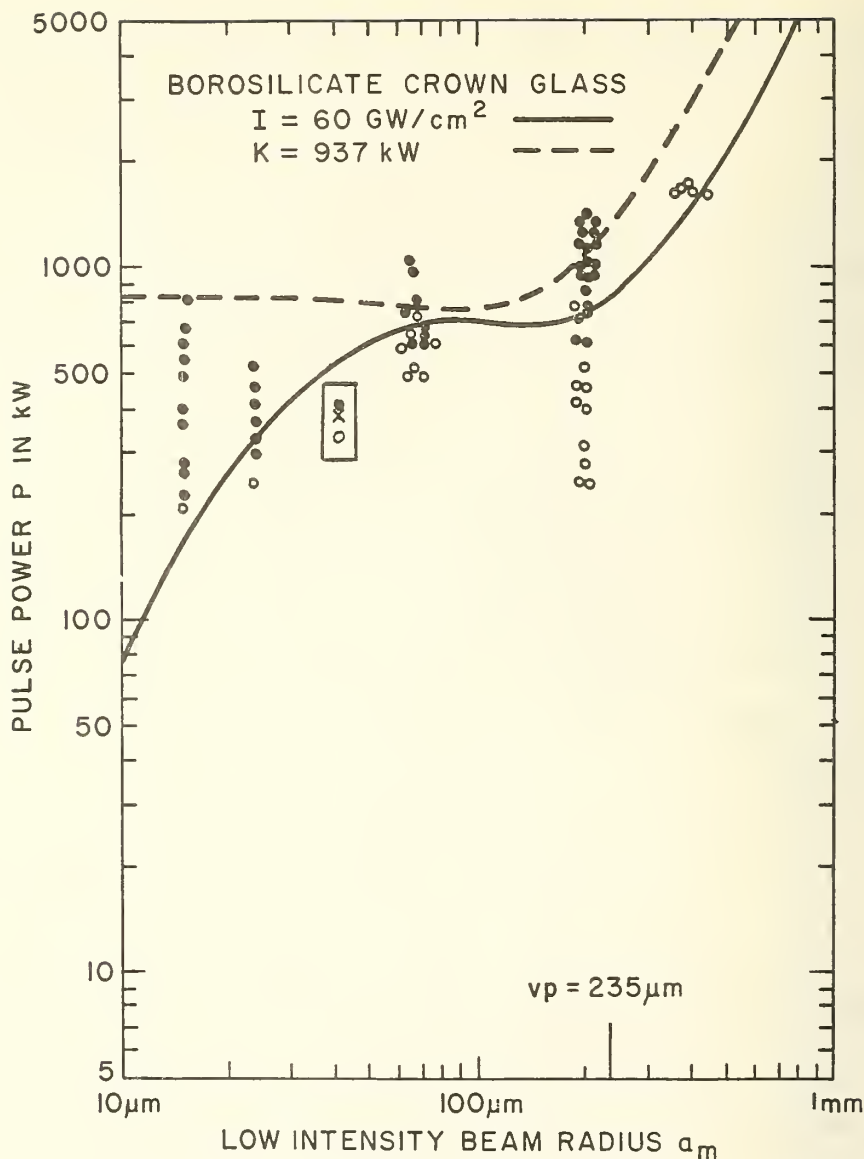


Figure 13. Track formation thresholds in BSC glass from the work of Kerr and of Steinberg (Ref. 9, 13). Also shown are data from the present work enclosed in the box. It was necessary to scale the data as the wavelength used and pulse width differed from Steinberg's. The open circles represent powers at which no damage occurred and the solid circles represent powers at which damage occurred. The X represents the scaled value of the critical power as obtained from Fig. 12.

$$P_c = \frac{\lambda^2 v^2}{(\rho_0 \frac{\partial n}{\partial \rho})^2} f\left(\frac{\tau v}{a_0}\right),$$

where v is the velocity of sound in the medium, $\partial n / \partial \rho$ is the change of refractive index with density, ρ_0 is the density of the material, and $f(\frac{\tau v}{a_0})$ is a known function. Thus the value of P_c which we obtain is scaled with wavelength and the value of a_0 is scaled with the pulse width. The wavelength dependence of $\rho_0 \partial n / \partial \rho$ is not taken into account as it is not known and is expected to be small. The result is that our data points compare favorably with Steinberg's. The result is important because a method now exists for matching self-focussing data taken with different wavelengths and different pulse widths.

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